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In-situ Capability of Ion Beam Modification and Characterization of Materials at Los Alamos National Laboratory*

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Abstract

The capability of *in-situ* ion beam modification and characterization of materials developed at Los Alamos National Laboratory is described. A beam-line from a 3 MV tandem accelerator and a beam-line from a 200 kV ion implanter are joined together in an *in-situ* target chamber. The chamber is equipped with a cold and hot sample stage with a temperature range from -100 to 500 °C. The angular (sample spin and basal rotation) motions and translational motions of the sample stage are controlled by a multi-axis goniometer. This chamber provides a unique capability to conduct a temperature dependent experiment of ion irradiation and sequential backscattering and channeling analysis. The efficiency and reliability of *in-situ* ion beam techniques are demonstrated by two examples, irradiation damage in (100) $MgAl_2O_4$ spinel crystals and ion-beam-induced densification of zirconia sol-gel thin films.

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1. *In-situ* ion beam techniques

In this paper, we describe a unique *in-situ* capability of combining ion beam analysis with ion beam modification, developed at the Ion Beam Materials Laboratory (IBML) at Los Alamos National Laboratory. The detailed description of ion beam facilities at the IBML has been published in Ref. 1 and this paper will only focus on the *in-situ* perspective of the facilities. Figure 1 shows a schematic diagram of the *in-situ* target chamber, also called the surface modification chamber. The chamber, in a dimension of 0.3 m diameter and 0.25 m height, is connected with beam-lines of a 200 kV ion implanter used for surface modification and a 3MV tandem accelerator used for surface characterization. The chamber is equipped with a multi-axis goniometer and a cold and hot sample stage. The angular motions (sample spin and basal rotation) of the goniometer are computer controlled. The temperature of sample stage can be varied from -100 to 500 C via liquid nitrogen cooling and resister heating. Two surface barrier detectors are installed in the chamber for the measurements of backscattering events (RBS). The freedom of varying incident and scattering geometry allows ion channeling and glancing geometry measurements. The irradiation beam spot on the target after rastered is about 6 mm diameter and the analytical beam of 2 mm diameter is centered in the irradiation spot. The charge integration of the irradiation beam is taken from a four-corner-cup charge-collector located on the 200 kV beam-line near the chamber. The charge of analytical beam is directly collected from the target. The capability of irradiation and sequential analysis provides an efficient and reliable method to conduct temperature dependent experiments including ion channeling, irradiation damage, and ion-beam-induced epitaxial regrowth for various materials. Two examples of using *in-situ*

capability, radiation damage in $MgAl_2O_4$ and ion-beam-induced densification of sol-gel zirconia films, are given below.

2. Radiation damage in $MgAl_2O_4$

Radiation effects in $MgAl_2O_4$ (spinel) has been a research subject of interest because of its radiation resistance and potential applications in radioactive environments. A study of the temperature effect on radiation damage in single crystal spinel has been conducted recently using the *in-situ* ion beam facilities at the IBML [2]. For sequential analysis, a 2 MeV He ion beam from the tandem was first aligned along the (110) axis of a (100) spinel wafer prior to irradiation. Then the sample was brought to a desired temperature for irradiation. Following each irradiation with an incremental dose using 370 keV Xe^{2+} ions from the ion implanter, an aligned RBS spectrum was sequentially taken from the same spot. Figure 2a shows examples of sequential channeling spectra taken from a spinel sample irradiated to various doses up to $8x10^{15}$ Xe/cm^2 at 120 °C. In comparison to the unirradiated spinel, the radiation-induced damage rendered in spinel increases with increasing irradiation dose. However, the amount of damage does not reach a fully random level up to a dose $2x10^{16}$ Xe/cm^2 (spectrum not shown) at which the estimated peak damage level is about 50 displacements per atom (dpa). The amount of damage retained in the Al and Mg cation lattice integrated through the irradiated layer [2] has been extracted from the channeling spectra in fig. 2a and plotted in fig. 2b as a function of irradiation dose. These channeling results reveal that spinel retains crystallinity under ion irradiation at 120 °C up to 50 dpa. This is in strong contrast to the ion irradiation at -170 °C where amorphization was found at 25 dpa [3]. Based on series studies on temperature effect, a radiation accumulation model in spinel is currently under development at the IBML.

3. Ion-beam-induced densification of zirconia sol-gel films

Densification of zirconia sol-gel films induced by ion-beam has been found recently by Levine et al. [4,5]. However, the initial conditions and non-uniformity of films depending on the sol-gel processing significantly affect the rate of densification and thus complicate the quantitative understanding of ion-beam-induced densification. Therefore, the *in-situ* analysis on the same sample spot provides a reliable way to limit the variation of experimental parameters. Figure 3a shows RBS spectra of sol-gel zirconia films coated on Si(100) wafers before and after 280 keV Ne ion irradiations at -100 °C to various doses up to $3x10^{16}$ Ne/cm^2. The increase in the height and the decrease in the width of the Zr RBS signal coincide with the densification of zirconia films due to the loss of H, C, and O. Figure 3b shows the experimental data of relative change in Zr RBS height as a function of Ne dose. The experimental data can be well fitted by a combination of two exponential terms. By further analyzing the change of the Zr height, and the change of the amount of retained H, C, and O in the films, we expect that the mechanisms involved in the ion-beam-induced densification will be explained.

4. Summary and future plans

The efficient and reliable *in-situ* ion beam capability provides a powerful and unique method to modify and characterize the material surface sequentially. This has been demonstrated through two examples recently being performed at the IBML, radiation damage of spinel and ion-beam-induced densification of sol-gel zirconia thin films. We are in the process of adding the analytical capability of both energy dispersive x-ray diffraction (at glancing and non-glancing angles) and particle induced x-ray emission into the *in-situ* chamber for detecting new phase

formation and trace elements. An excimer laser will be coupled to the chamber for laser-solid interaction studies.

References

[1] J.R. Tesmer, D.M. Parkin, and C.J. Maggiore, Mater. Res. Soc. Bull. XII (1987) 101.

[2] N. Yu, M. Nastasi, M.G. Hollander, C.R. Evans, C.J. Maggiore, K.E. Sickafus, J.R. Tesmer, Mat. Res. Soc. Symp. Proc. 316 (1994) 69.

[3] N. Yu, K.E. Sickafus, and M. Nastasi, Philos. Mag. Lett. 70 (1994) 235.

[4] T.E. Levine, J.L. Keddie, P. Revesz, J. W. Mayer, and E.P. Giannelis, J. Am. Ceram. Soc. 76 (1993) 1369.

[5] T.E. Levine, P. Revesz, E.P. Giannelis, and J. W. Mayer, J. Vac. Sci. Technol. B 12 (1994) 986.

Figure captions

Figure 1 Schematic drawing of top view of the *in-situ* surface modification chamber connected with a beam-line of 3MV tandem accelerator and a beam-line of 200 kV ion implanter.

Figure 2 (a) Aligned backscattering spectra from single crystal $MgAl_2O_4$ irradiated sequentially with 370 keV Xe ions at 120 °C, along with a random spectrum; (b) damage fraction of Al and Mg cation sublattice as a function of Xe dose.

Figure 3 (a) Backscattering spectra from sol-gel zirconia thin films on Si (100) wafers irradiated sequentially with 280 keV Ne ions at -100 °C; (b) relative increase in Zr RBS height with increasing Ne dose.

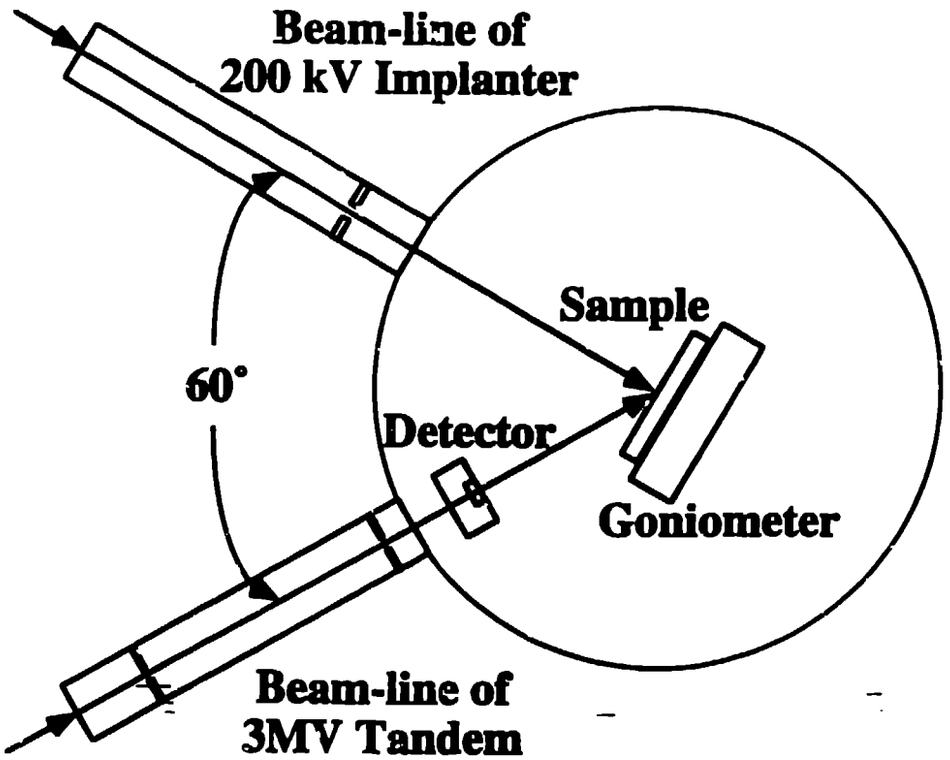

Beam-line of
200 kV Implanter
Sample
60°
Detector
Goniometer
Beam-line of
3MV Tandem

Figure 1

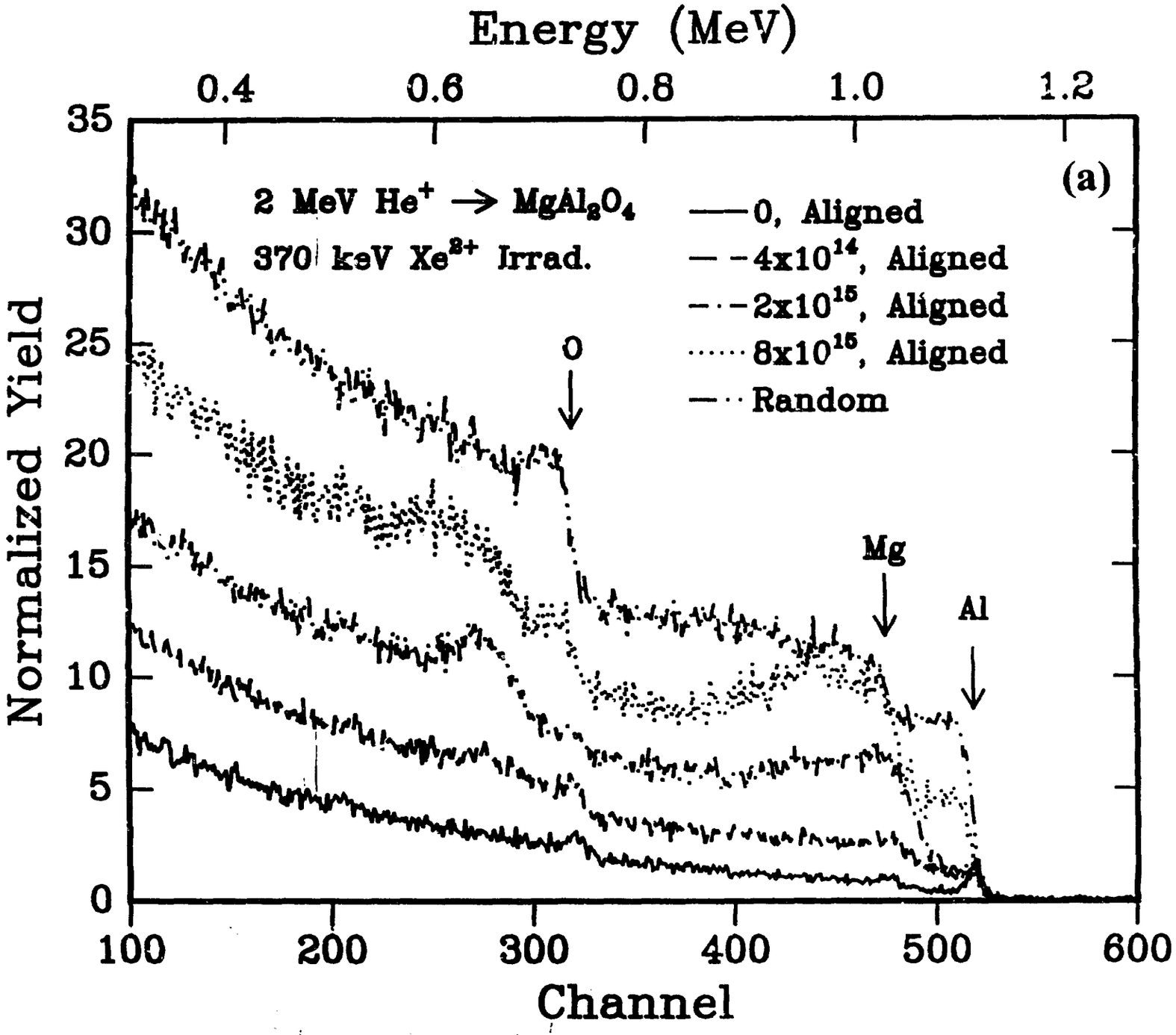

Energy (MeV)
0.4
0.6
0.8
1.0
1.2
35
30
25
20
15
10
5
0
Normalized Yield
(a)
2 MeV $He^+ \rightarrow MgAl_2O_4$
370 keV Xe^{2+} Irrad.
0, Aligned
$4x10^{14}$, Aligned
$2x10^{15}$, Aligned
$8x10^{15}$, Aligned
Random
O
Mg
Al
100
200
300
400
500
600
Channel

Figure 2a

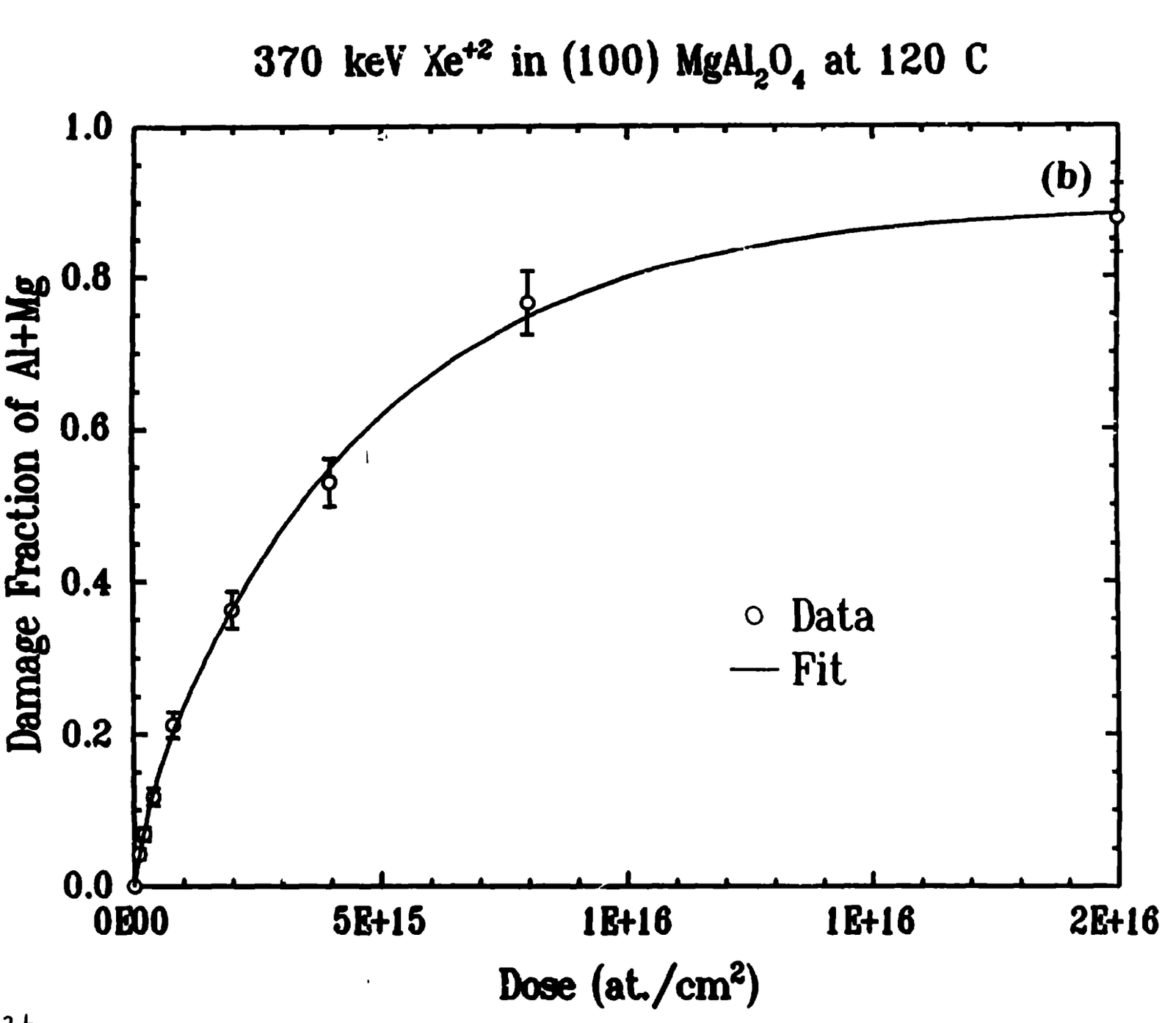

370 keV Xe+2 in (100) MgAl2O4 at 120 C
(b)
Damage Fraction of Al+Mg
1.0
0.8
0.6
0.4
0.2
0.0
Data
Fit
0E00
5E+15
1E+16
1E+16
2E+16
Dose (at./cm2)

Figure 2b

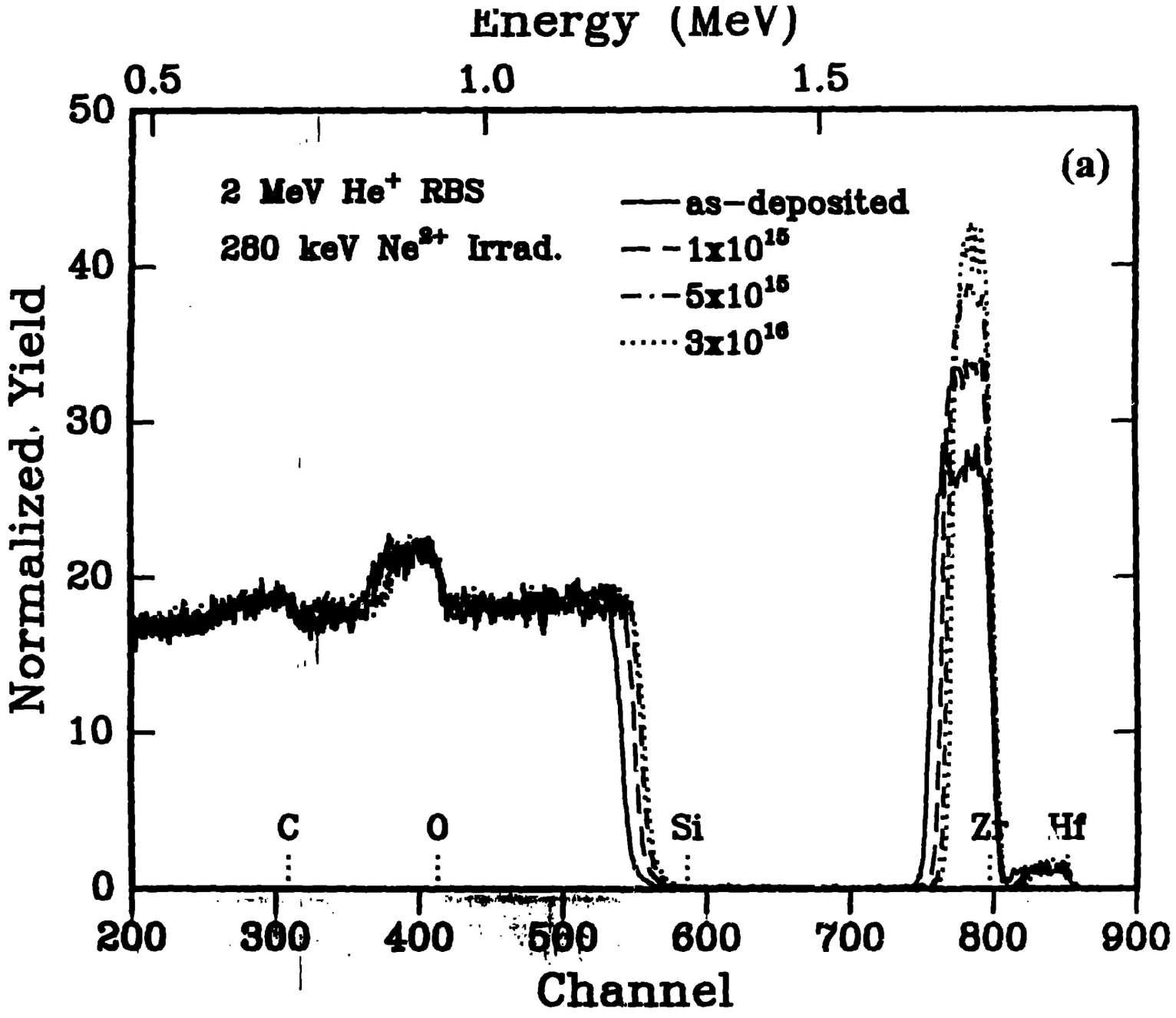
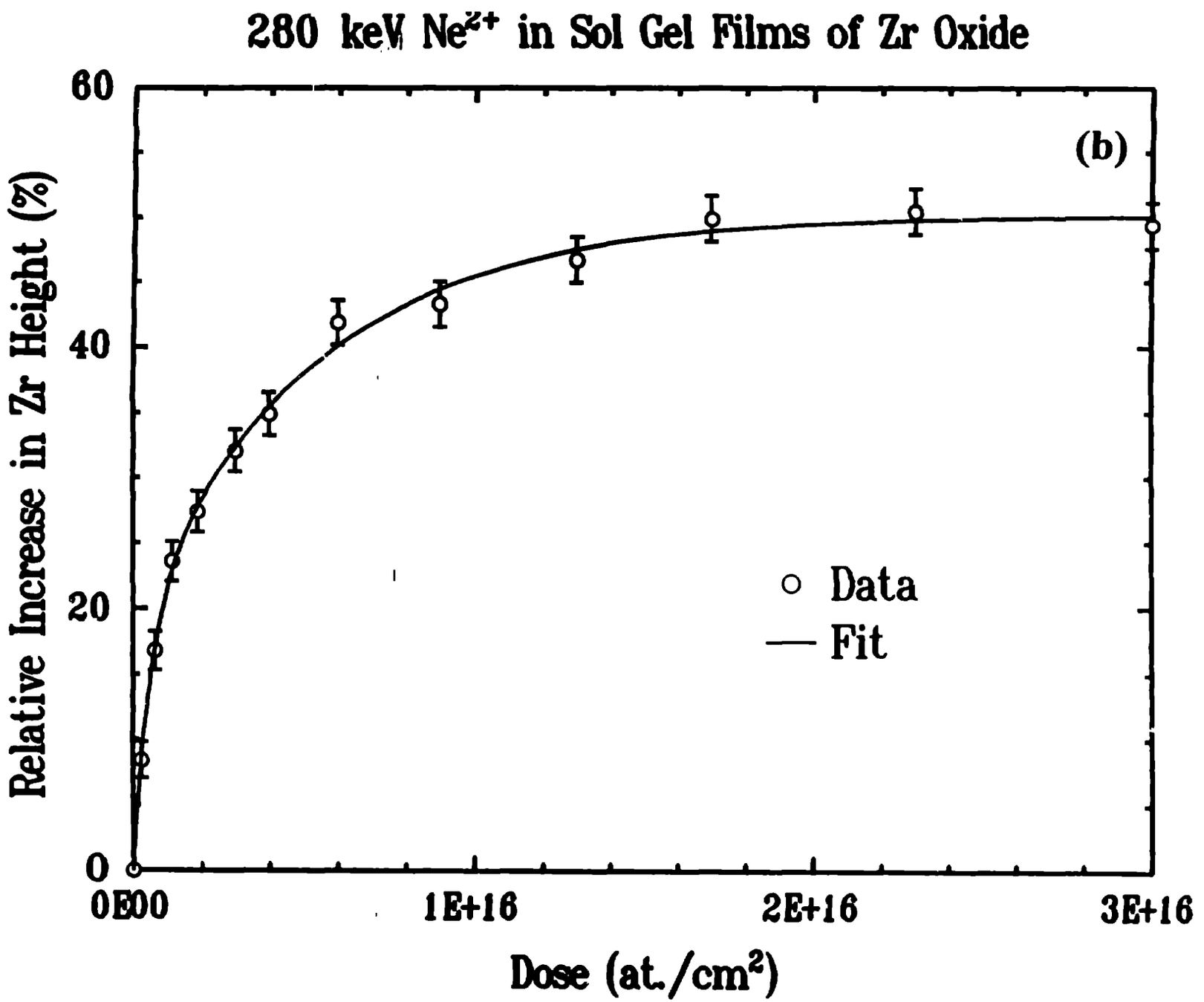

Energy (MeV)
0.5
1.0
1.5
(a)
2 MeV He^+ RBS
280 keV Ne^{2+} Irrad.
as-deposited
$1x10^{15}$
$5x10^{15}$
$3x10^{16}$
Normalized. Yield
0
10
20
30
40
50
C
O
Si
Zr
Hf
200
300
400
500
600
700
800
900
Channel

Figure 3a


280 keV Ne^{2+} in Sol Gel Films of Zr Oxide
(b)
Relative Increase in Zr Height (%)
60
40
20
0
0E00
1E+16
2E+16
3E+16
Dose (at./cm²)
Data
Fit

igure 3b